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TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. § 371		Attorney's Docket Number 045636-5057
International Application. No.	International Filing Date	U.S. Application No. Unassigned <b>10/089176</b>
PCT/FR00/02664	September 27, 2000	Priority Date Claimed September 27, 1999

Title of Invention: **METHOD AND DEVICE FOR ATOMIC INTERFEROMETRY NANOLITHOGRAPHY**

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Applicants herewith submit to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. § 371.
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. § 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. § 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. § 371(b) and PCT Articles 22 and 39(I).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. § 371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. § 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. § 371(c)(3)).
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. § 371(c)(3)).
9. ☐ An oath or declaration of the inventors (35 U.S.C. § 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. § 371(c)(5)).

**Items 11. to 14. below concern other document(s) or information included:**

11. ☐ An Information Disclosure Statement under 37 C.F.R. § 1.97 and § 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. § 3.28 and § 3.31 is included.
13. ☐ A FIRST preliminary amendment.
14. ☒ A SECOND or SUBSEQUENT preliminary amendment.
14. ☒ Other items or information:
  - a. WO 01/24219 cover sheet including English language abstract
  - b.
  - c.
  - d.
  - e.
  - f.

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b. ☒ **Except** for issue fees payable under 37 C.F.R. § 1.130, the Commissioner is hereby authorized by this paper to charge any additional fees during the entire pendency of this application including fees due under 37 C.F.R. § 1.16 and § 1.17 which may be required, or credit any overpayment to Deposit Account No. 50-0310.

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**A METHOD AND AN INSTALLATION FOR PERFORMING  
NANOLITHOGRAPHY BY ATOMIC INTERFEROMETRY**

The invention relates to a method of performing lithography at nanometer scale by using atomic  
5 interferometry, and it also relates to an installation for implementing such a method.

**BACKGROUND OF THE INVENTION**

The invention relates to the field of lithography at sub-micron scale. By projecting beams of atoms either of  
10 alkali type (e.g. Na, Li, or Cr) or of metastable type (He\* or Ar\*), atomic lithography makes it possible, via a mask, respectively to cause substance to be deposited on a substrate to be treated or to cause a pattern to be etched in a resin deposited on that substrate. The  
15 article by M. Kreis et al., published in Applied Physics, Vol. B63, 649 (1996), illustrates that type of technique.

Compared with the more conventional technique of photon lithography, atomic lithography presents advantages which relate to the implementation conditions  
20 and to the physical limits of these techniques:

- the photon source, generally a UV laser, requires high brightness and means that are complex and expensive for producing photons at shorter and shorter wavelengths in order to increase the resolution of the installation  
25 (e.g. an Nd:YAG type laser for exciting a supersonic jet of xenon atoms);

- a magnifying optical system (magnifying by a factor of 4 or 5) formed by multilayer mirrors of reflectivity that is selective in wavelength and of  
30 limited lifetime; and

- pattern thickness is limited by the wavelength used, e.g. 157 nanometers (nm) for the above-mentioned xenon jet in devices for producing extreme UV radiation close to soft X-rays.

35 Micro-lithographic techniques based on atomic optics use thermal or quasi-supersonic beams of atoms that are confined in a magneto-optical trap. In such techniques,









- Figure 4 is a perspective view of an example of how the interference pattern can be moved in translation by adding a uniform magnetic field;

- Figures 5a to 5d are interference profiles of an  
5 installation of the invention as obtained under various operating conditions; and

- Figure 6 shows examples of interference background around the central spot being reduced by using multiple interferometers.

#### 10 MORE DETAILED DESCRIPTION

The nanolithography installation shown in Figure 1 is based on a Stern-Gerlach type interferometer. The component elements of such an interferometer comprise in succession, along a central axis Z'-Z, a polarizer 3, a  
15 set of bars 51 producing a transverse magnetic field forming the phase object between two separator plates 4 and 6, followed by analyzer 7 and then a detector 8 which is located at the position on the substrate where deposition or etching is to take place.

20 The atomic interferometer receives a beam of metastable helium atoms  $\text{He}^*$  delivered by a source 1. The source has a continuous electrical discharge triggered in expansion through a nozzle between a conical tungsten electrode and the nozzle. The discharge takes place at a  
25 voltage of about 1 kiloelectron volts (keV) and at a current of several milliamps (mA). Advantageously, the discharge can be pulsed and the resulting gas can be cooled, e.g. using liquid nitrogen.

The density at which atoms are delivered is then  
30 greater than about  $4 \times 10^{14}$  atoms per second per steradian (atom/s/srd) over an angular range of 0.5 radians (rd), and the speed distribution of the atoms is of the Maxwell type, and is broad, being about 30% about a mean value of 2 km/s. By superposing interference patterns having  
35 different pitches, such dispersion give rise to interference rings with contrast that falls off quickly. As shown in Figure 2, in the absence of any dispersion in



speed, the interference pattern  $F$  presents constant contrast between bright interference fringes  $I_b$  and dark interference fringes  $I_s$ . However with speed dispersion of about 30%, associated with Maxwell type distribution, only the bright central fringe  $I_c$  forming a fine central spot remains, as can be seen in Figure 3, which spot stands out significantly from the other rings which are highly attenuated along the  $Z'-Z$  axis. Figures 2 and 3 are simulations which take account of all of the parameters that are quantified in the present description.

The angular aperture of the helium jet output by the source 1 is defined by the collimation performed by transverse cooling. This cooling is implemented by means of two optical molasses 2a and 2b acting on the transverse speed components of the atoms, respectively along the  $X'-X$  axis and along the  $Y'-Y$  axis. They use two laser beams 2f with red-shifted broadened frequency side bands of width 15 MHz to 20 MHz, the spectral offset and width being obtained using an acousto-optical modulator (not shown). These laser beams are circularly polarized. The beams 2f are subjected to a series of reflections between two facing plane mirrors  $M$  located to form a "zigzag" of width equal to 8 millimeters (mm), the interaction length then being raised to 8 centimeters (cm) for each molasses. The working laser power remains low, about 40 milliwatts (mW). Under such conditions, the beam of atoms presents an aperture of about 0.1 rd and transverse speeds of less than 0.1 m/s.

The optical interaction between the beam of helium atoms and the polarizer 3 selects a Zeeman sublevel relative to the quantization axis of the polarizer (with magnetic field) implemented by light pumping in one embodiment. This pumping is obtained by means of a laser beam polarized by a distributed Bragg reflector (DBR) diode having a wavelength of 1.08 micrometers ( $\mu\text{m}$ ) and



The installation makes it possible to obtain a gradient  $G$  of about  $5 \times 10^{-4} i_A/\text{mm}$ , but greater values, e.g. those used for simulations, of the order of  $10^{-2} i_A/\text{mm}$  can easily be obtained by using other devices, e.g. coils in an anti-Helmholtz configuration or electromagnets in a hexapolar configuration.

The diameter of the central spot becomes finer with increasing gradient  $G$ . The installation gives gradients that are relatively modest, but it enables a spot to be obtained of a size that is about 10 nanometers. The gradient  $G$  is kept constant in the vicinity of the axis  $Z'-Z$  which is of a size similar to that of the pattern to be etched, e.g. a few micrometers. Only the initial width of the profile of the beam of atoms leaving the source is large relative to the size of the pattern to be etched or to be deposited.

The second separator plate 6 is made and operates in the same manner as the first separator plate 4 to form coherent superpositions for each atomic state emerging from the phase object. Other techniques exist for forming such a separation, e.g. diffraction through physically-embodied gratings having ultrafine slits, e.g. at 20,000 lines per millimeter, or optical gratings formed by a standing wave produced by laser radiation reflected on a mirror or by the reversal effect obtained by absorbing a resonant photon.

The analyzer 7 retains only a single Zeeman state so that the emerging flux of atoms contains only a series of interference terms forming annular bright and dark fringes of intensity that is measured by the detector 8. In the embodiment described, the analyzer 7 is constituted using a laser beam of the same type as that which forms the polarizer 3. This detector is substituted by the silicon substrate coated in resin which is to be etched or on which the desired structure is to be deposited.



The corresponding conditions are summarized in the table below, as a function of: source width ( $\sigma$ ); of relative width of the longitudinal speed distribution of the beam ( $\delta v/v$ ); of the diameter of the resulting central spot ( $\delta x$ ); and of the distances of the main components of the interferometer ( $d$  = distance between the source and the phase object,  $L$  = the width of the phase object, and  $D$  = the distance between the detector/substrate and the source).

10 TABLE

Fig.	$\sigma(\mu\text{m})$	$\delta v/v$	$d(\text{cm})$	$L(\text{cm})$	$D(\text{cm})$	$G/\mu\text{m}$	$\delta x(\text{nm})$	Sources
5a	0.2	1	5	5	11	$4 \times 10^{-6}$	40	coherent
5b	2000	2	5	20	200	$4 \times 10^{-7}$	100	coherent
5c	500	1	5	10	25	$8 \times 10^{-7}$	80	incoherent Zeeman state $M=0$
5d	2000	1	5	10	100	$4 \times 10^{-7}$	120	coherent Zeeman state $M=0$

These examples show that spot diameters of one to a few tens of nanometers can easily be achieved under normal conditions. It should be observed that departures from the ideal shapes shown in Figures 5a to 5d give rise to deformation of the interference pattern but that this deformation is of no significance in any event.

It should be observed that the intensity of the emitted atomic current, i.e. the number of atoms per second and per square centimeter is equal to the intensity of the current on the resulting central spot, since the modulation implemented by the gradient acts only on the background intensity which is  $2J+1$  times smaller, where  $J$  is the spin of the atom used. For example, for argon ( $J=2$ ), the background is five times

smaller, and for a source emitting at  $4 \times 10^{14}$  atoms/s/srd a current density of  $1.3 \times 10^{10}$  atoms/s.cm<sup>2</sup> is obtained when the phase object distance D is equal to 0.5 m.

5 It should also be observed that in the etching or deposition mechanism, it is the internal energy of the metastable atoms used which performs the main function rather than the kinetic energy thereof, since the effect of speed is negligible.

10 For argon, the internal energy is about 15 eV per atom transferred to the resin, for helium the internal energy is greater than or equal to 20 eV. This energy is quite sufficient for etching a polymer film.

15 In the deposition process, the speed of the atoms is limited and the atom reflection factor becomes negligible. This applies to thermal speeds for "deposable" atoms of the alkali type or chromium.

20 In any event, it is the number of atoms reaching the target during a determined time interval which needs to be taken into consideration and not the speed of the atoms. By way of example, a threshold density can be about  $10^{14}$  atoms/s/srd. In particular, the "deposable" atoms present a flux that is considerably greater (about  $10^4$  times greater) than that of the metastable atoms, leading to a current density of about  $10^{14}$  atoms/s.cm<sup>2</sup>.

25 For etching, the metastable atoms do not dig directly into the resin covering the silicon medium, but they modify its properties by breaking its molecules with efficiency approaching unity. For example, several tens of atoms reaching an area having a diameter of 40 nm produce the desired effect which takes about 60 s. For deposition purposes, in order to deposit a thickness of 1 nm on the same area, it is necessary to deposit  $10.5 d/M_A$  where  $d$  is the density and  $M_A$  is atomic mass, and this requires a duration of a few milliseconds.

35 The atomic spot appears on a background of uniform intensity which is equal to  $1/(2J+1)$  of the intensity of the central spot. When the tracing speed is relatively

low (about 0.7 nm/s), accumulated background intensity can become a drawback. This disturbance can be avoided by using a multiple interferometer, using a succession of interferometers placed in series, the analyzer of one  
5 interferometer being used as the polarizer of the next interferometer. Figure 6 shows the final transverse profile as modulated by the successive interferometers when the number  $n$  of successive identical interferometers in series varies over the range 1 to 4. Contrast is thus  
10 very significantly improved since if  $M(\rho)$  designates the modulation induced by each interferometer, the final transverse profile is  $M(\rho)^n$ . The half-height diameter of the central spot is also reduced, by a statistical factor equal to  $\sqrt{n}$ .

15 The invention is not limited to the embodiments described and shown. The relationship describing the transverse distribution of density of atoms in the beam can be of the Gaussian type, for example, with a standard deviation of 100  $\mu\text{m}$ .

## CLAIMS

- 1/ A method of lithography by atomic interferometry on a target, on the basis: of spin polarizing a beam of incident atoms by optical pumping; of forming a phase object by transverse magnetic induction on the basis of a coherent superposition of spin states; and then of analyzing the beam of atoms by optical interaction so as to retain only a single spin state; the emerging beam of atoms containing a series of interference terms; wherein the magnetic induction presents a transverse gradient to form an annular interference pattern, wherein the beam of atoms presents a speed distribution greater than 20% in order substantially to eliminate interference fringes other than the central fringe which then forms a spot, and wherein adjustable transverse uniform magnetic induction is added to the induction having a gradient so as to move the central spot in translation in predetermined manner over the target.
- 2/ A method of lithography according to claim 1, in which the gradient of the transverse induction is adjusted as a function of the desired spot intensity and diameter, the resulting spot becoming finer and more intense with increasing gradient.
- 3/ An installation for nanolithography by atomic interferometry, the installation comprising a Stern-Gerlach type interferometer with a phase object in the form of four-pole magnetic induction with a transverse gradient created by four parallel bars carrying alternating direct currents, bracketed between two separator plates, preceded and followed respectively by a spin polarizer and by an analyzer operating by laser pumping, the additional uniform field being created by four other additional bars fed in paired manner with adjustable currents in order to create a uniform field of



arbitrary orientation and intensity by adjusting two parameters.

4/ An installation for nanolithography by atomic  
5 interferometry according to claim 3, in which the  
additional bars have the same length as the main bars,  
are disposed at  $45^\circ$  relative thereto, and carry paired  
currents.

10 5/ An installation for nanolithography according to claim  
3, in which the additional field bars comprise two pairs  
of coils in Helmholtz configuration.

6/ An installation for nanolithography by atomic  
15 interferometry according to claim 3, in which the source  
of atoms is a source for continuously discharging  
metastable helium or argon with an approximately Maxwell  
type speed dispersion of about 30% to 40% around  
approximately 2 km/s for atoms of helium and 500 m/s for  
20 atoms of argon.

7/ An installation for nanolithography by atomic  
interferometry according to claim 3, in which the laser  
pumping of the polarizer is performed by a circularly  
25 polarized laser diode, the spin polarization being  
performed on Zeeman level +1 or -1.

8/ An installation for nanolithography by atomic  
interferometry according to claim 3, in which the  
30 analysis is performed by deflecting atoms that lie in  
Zeeman states other than the selected state by using  
different light frequencies by means of at least one  
acousto-optical modulator coupled to the laser in the  
presence of a magnetic field that is intense, e.g. of the  
35 order of 100 G.

- 9/ An installation for nanolithography by atomic interferometry according to claim 3, in which the separator plates are made up of physically-implemented gratings with ultrafine slits, or of optical gratings formed by a standing wave produced by laser radiation reflected on a mirror, or else by means of a very low intensity magnetic field turning through  $90^\circ$  to induce transitions between the Zeeman states.
- 10/ An installation for nanolithography by atomic interferometry according to claim 3, in which the beam of atoms is collimated by transverse cooling by means of a two-dimensional optical molasses made up of two successive identical molasses acting respectively on one or the other of the transverse components, using laser beams that are laterally enlarged in frequency, being shifted towards the red by means of an acousto-optical modulator and circularly polarized, and each molasses is formed by a series of "zigzag" reflections of the laser beam on two facing plane mirrors.
- 11/ An installation for nanolithography by atomic interferometry according to claim 3, in which the gradient of the transverse magnetic field is created by two coils in an "anti-Helmholtz" configuration, or by a set of electromagnets placed in a multipolar or  $2n$ -polar configuration.
- 12/ An installation for nanolithography by atomic interferometry according to claim 3, in which the source makes use of a discontinuous electrical discharge struck in expansion through a nozzle between a conical electrode made of tungsten and the nozzle, thereby forming jets of atoms either of the alkali or chromium type, or else of the metastable hydrogen or inert gas type to obtain respectively deposition on a substrate that is to be

treated or etching of a pattern in a resin placed on the substrate.

13/ An installation for nanolithography by atomic  
5 interferometry according to claim 3, in which the  
interferometry is multiple in that use is made of a  
succession of interferometers in series, the analyzer of  
one interferometer being used as the polarizer of the  
next in order to eliminate the background from the  
10 interference pattern and in order to refine the central  
spot.

## A B S T R A C T

The invention proposes a novel technique for implementing high performance atomic lithography, and in particular high resolution lithography. The technique makes use of Stern-Gerlach type atomic interferometry enabling disturbances to be implemented in the atomic phase of the beam. Such interaction then directly modulates the intensity of the associated wave in the plane extending transversely to the beam of atoms, and does so in controllable manner. The installation of the invention for nanolithography by atomic interferometry comprises a Stern-Gerlach type interferometer comprising, as its phase object, four-pole magnetic induction having a transverse gradient created by four parallel bars carrying alternating direct currents, bracketed between two separator plates, preceded and followed respectively by a spin polarizer and by an analyzer operating by laser pumping. An additional uniform field is being created by another four additional bars powered in paired manner by adjustable currents in order to create a uniform field of arbitrary intensity and orientation for the interference pattern by adjusting the two current parameters. The source of atoms is a source that continuously discharges metastable helium or argon with approximately Maxwell type speed dispersion of about 30% to 40% in order to obtain a central spot.

**COMBINED DECLARATION FOR PATENT APPLICATION AND POWER OF ATTORNEY**U.S. DEPARTMENT OF COMMERCE  
Patent and Trademark Office

ATTORNEY DOCKET NO. :

As a below named inventor, I hereby declare that :

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled :

**METHOD AND DEVICE FOR ATOMIC INTERFEROMETRY NANOLITHOGRAPHY**

the specification of which :

is attached hereto ; or

was filed as United States application Serial No. \_\_\_\_\_ on \_\_\_\_\_ and was amended on \_\_\_\_\_ (if applicable) ; or

was filed as a PCT international application Number PCT/FR00/02664 on September 27, 2000 and was amended under PCT article 19 on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose the U.S. Patent and Trademark Office information which is material to the patentability of claims presented in this application in accordance with Title 37, Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate or §365(a) of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed :

**PRIOR FOREIGN APPLICATION(S) :**

COUNTRY (if PCT, indicate PCT)	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED
FRANCE	99/12016	27 September 1999	X



Combined Declaration For Patent Application and Power of Attorney – (Continued)  
(includes Reference to PCT International Applications)

ATTORNEY DOCKET NO. :

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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Listing of Inventors Continued on attached page(s) [ ] Yes [ X ] No